UCRL- 95645
PREPRINT

# The Effect of Aging on Smoke Optical Properties and Scavenging Characteristics

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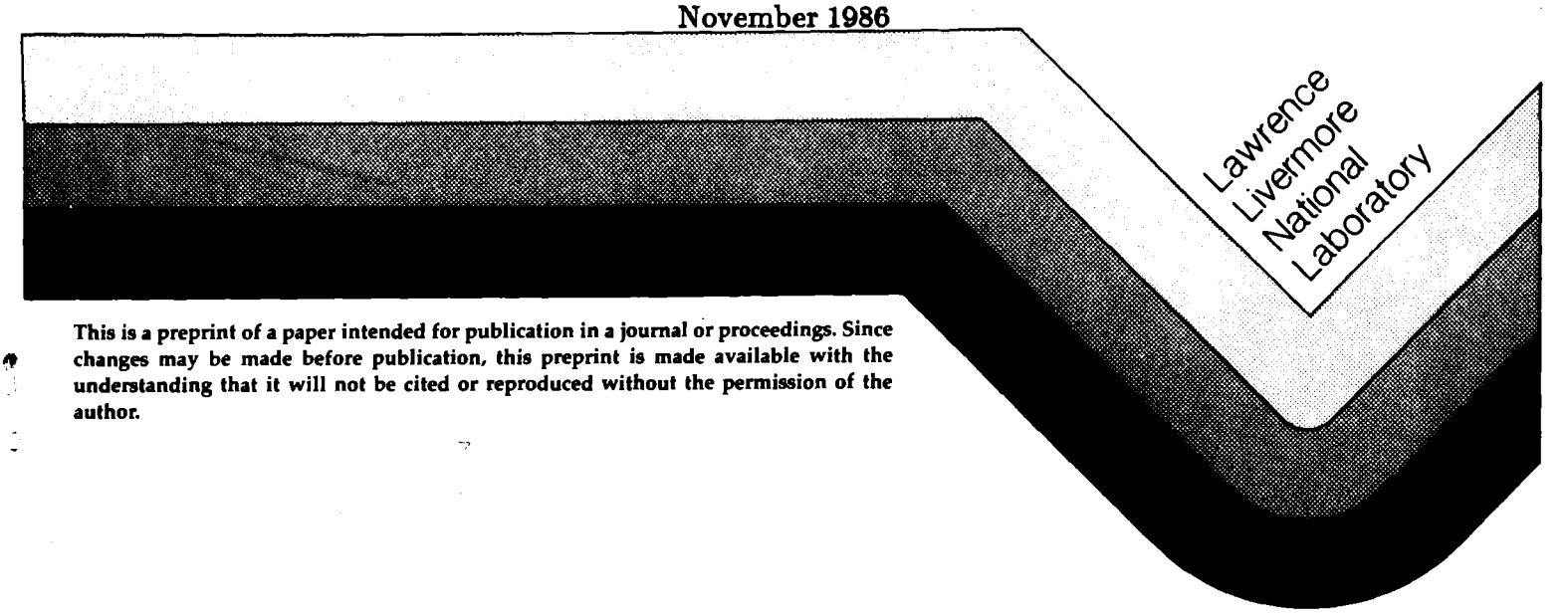
This paper was prepared for the
DNA workshop on Smoke Emission and Properties
November 13–14, 1983, held at
National Bureau of Standards
Gaithersburg, MD

November 1986



## DISCLAIMER

# The Effect of Aging on Smoke Optical Properties and Scavenging Characteristics

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Abstract

Aging of smoke in dense smoke plumes is known to alter its size distribution and scavenging characteristics. In this paper, I review data pertaining to these processes and compare them to model simulations of the coagulation of smoke. Model simulations of the degree of smoke coagulation in the first few days after a nuclear war are summarized. The predicted size of smoke particles after several days of coagulation is found to be larger than that of any data pertaining to the absorption properties of smoke. Thus, it is suggested that more relevant data on the absorption properties of smoke is needed.

I also review aging experiments pertaining to the number of cloud condensation nuclei in a smoke sample. I show that the fraction of smoke particles which act as CCN after aging depends on the number of particles initially present in the aging chamber. Smoke from an acetylene flame can quickly coagulate to sizes wherein nearly all of the particles act as CCN. On the other hand, only 10 percent of the smoke particles from an outdoor fire of gasoline and diesel fuel became CCN after 30 hours of aging. The development of CCN concentrations in this experiment may have been quenched by low initial concentrations in the aging chamber. Both experiments are consistent with particles as small as 0.08 micron in radius (and perhaps even smaller) acting as CCN. Model simulations of the coagulation of smoke particles above a large, intense fire show that coagulation would allow approximately 50 percent of the particles to become larger than 0.08 micron before the plume reaches cloud base. Furthermore, aging over several days time would transform nearly all the particles into the scavengable size range.

## 1. Introduction

Several processes may act to alter smoke particles as they age. Coagulation can alter their size distribution. Gas-to-particle conversion and chemical reactions can alter their chemical nature. Reaction with ozone may even destroy smoke particles. The degree to which these processes alter the smoke characteristics depends on the time scales over which aging is considered and the local environment of the particles (e.g. the smoke and gas concentrations). In this paper, I shall review what is known about aging by coagulation. Aging of smoke particles is important to the evaluation of the effects of post-war smoke on the environment because changes in the chemistry and size distribution of smoke particles may alter the optical properties of the smoke as well as its propensity for removal by either precipitation or deposition. We therefore also discuss how aging might alter the absorption and scattering properties of smoke and its rate of removal by precipitation and deposition.

## 2. Coagulation of smoke particles

Smoke particles are known to change their size characteristics by coagulation. This may occur on relatively short time scales within a dense smoke plume (Radke, et al., 1980; Hallett et al., 1986) or on longer time scales, if concentrations remain high (Turco et al., 1983; Crutzen et al., 1984; Penner and Porch, 1986). In addition, if large amounts of dust and ash are lofted in high-wind firestorms, significant coagulation on short time scales may occur (Porch, et al., 1985). This latter phenomena, however, is not important except under special circumstances whose frequency is not likely to be large (Pittock, et al., 1986; Turco et al., 1986; Porch et al., 1986).

Model results for degree of coagulation.

Three estimates of the effects of coagulation on smoke properties have been made in addition to that in the National Academy of Sciences report (Turco et al., 1983; Crutzen et al., 1984; NRC, 1985; and Penner and Porch, 1986). Turco et al. (1983) estimated that dispersion of the smoke plumes over eight months time to hemispheric scales might cause enough coagulation to reduce the average opacity after 3 months by 40 percent. This estimate was increased to 75 percent when the adhesion efficiency of the particles was maximized. However, no information on the initial concentration of particles or their size distribution was given, so that it is difficult to compare this estimate with those of others. Crutzen et al. (1984) included the effects of coagulation for an assumed "self-preserving" size distribution with an initial mode radius of 0.2 micron. For an initial mass concentration of 5 mg $m^{-3}$, they calculate a moderate increase in the mode radius of the smoke size distribution from 0.2 micron initially to about 0.3 micron after 10 days. They apparently included the effects of this change on the optical properties of the smoke but this was not separately reported. The writers of the NAS report concluded that if initial concentrations were less than about $10^6$ $cm^{-3}$, prompt coagulation would be unlikely to increase the mode radius of the smoke particle distribution above 0.2 to 0.3 micron. However, they did not consider time scales for coagulation that were longer than one day. Penner and Porch (1986) estimate the most significant size changes due to coagulation. Based on a three-dimensional model simulation of the dynamics associated with a large, intense fire, they argue that initial concentrations should be as high as 50 mg $m^{-3}$ and find that under these circumstances coagulation can create a mode radius near 0.4 microns radius after a few days coagulation. Significantly, the development of a mode near 0.4 micron is consistent with the observation of a "blue sun" after the great smoke pall caused by the Alberta forest fires in 1950 (Wexler, 1950).[1] Penner and Porch (1986) estimate that the specific absorption coefficient of the smoke could decrease by as much as a factor of 3 (or 68 percent) in about a week, for spherical smoke particles with a refractive index of m=1.75 -0.5i. Less absorbing smoke, with m=1.5 -.05i, would not significantly change its absorption properties although its scattering coefficient could at first increase and then decrease to about half its initial value in about a week. These estimates for coagulation and those of the authors previously cited, depend, of course, on the assumed initial smoke concentration and rates of spread of the smoke plumes. Since the absorption

[1] The development of a mode radius larger than 0.4 micron is also consistent with growth by condensation.

of solar radiation by smoke can alter circulation patterns and perhaps cause more rapid dispersion of the smoke plumes, these assumed rates should be verified by comparison with rates calculated in a mesoscale model that treats plume dynamics as the smoke spreads over the first several thousand kilometers.

Effects of coagulation on optical properties.

The effects of coagulation on the optical properties of spherical particles are described well by Mie theory. Thus, to the extent that the smoke particles become spherical, the theoretical results described above for the decrease in absorption and scattering properties of smoke as a result of aging may be considered reliable. However, if the particles remain in a chained-agglomerate morphology, we have little knowledge of the effects of coagulation on optical properties. Theory suggests that as chained agglomerates grow, they should increase their absorption per unit mass by perhaps a factor of two (Roessler, et al., 1981) or remain constant (Berry and Percival, 1986). However, these theoretical results are all based on assumptions that are only valid in the Rayleigh limit–i.e. for particles whose size is much less than the wavelength of light. Based on the theoretical predictions of Penner and Porch (1986), we need to understand the absorption properties of particles whose volume is equivalent to that of spheres with a radius of about 0.4 micron. If these particles survived the initial cloud and rainout processes and remained in a chained-agglomerate morphology, they would contain some 8000 primary spherical particles with radii near 0.02 micron. We may use the model results of Mulholland et al. (1986) to predict that the radius of gyration for such a particle would be about 2.4 micron.

Unfortunately, the absorption properties of such large particles are not known. The measurements obtained so far do not address such large particles. The largest particles considered in the study of Lee (1983), for example, had a lengthwise measurement of only 1 micron, and thus are five times smaller than the particles expected after a few days coagulation in the smokey post-war environment. Furthermore, even the studies of Lee (1983) are questionable because they were made using the integrating plate technique. In this technique the particles are first captured on a filter. The technique would therefore overestimate absorption to the extent that the large particles tended to lie flat on the filter as opposed to being randomly oriented. Most researchers using this technique believe that under most circumstances the particles are randomly distributed. However, this is more difficult to ensure for very large particles.

Only very sketchy data are available which suggests a decrease in absorption for very large particles. For example, Sutherland et al. (1984) measured the specific extinction coefficient for smoke from a diesel fuel fire. This smoke contained a large number of large carbon aggregates, but the size distribution was not specifically measured. The measured specific extinction coefficienct was only 5.6 $m^2/g$. The specific absorption coefficient was not measured, but could have been roughly half this value. Since the specific absorption coefficient for "fresh smoke" is 8–10 $m^2/g$, this measurement suggests that there can be a significant decrease in the specific absorption coefficient of large aggregates. Another experiment, by O'Sullivan and Ghosh (1973), showed that the optical density of smoke at 0.5 micron relative to that at 2.2 micron decreases when the particles are allowed to coagulate. This observation is also consistent with decreased absorption for large particles, but again the size distribution was not measured. Thus, unfortunately, until relevant data

to this size range become available, we can only estimate that the absorption for large agglomerate particles with physical dimensions of several microns should lie somewhere between the specific absorption of "fresh" smoke and that of large (0.4 micron radius) spheres.

Comparison of coagulation model to data.

Radke et al. (1980) show evidence of coagulation in the plume of the Meteotron. After 30 minutes, their data suggest that a mode radius near 0.1 micron is developing. Another peak is seen in the concentrations of the very smallest particles-those with radii less than 0.01 micron. A similar phenomena develops in the data of Hallett et al. (1986) with a mode developing between 0.1 and 0.3 micron radius after 30 hours. Also, particles with an apparent radius of less than 0.05 micron are still present in significant concentrations after 54 hours. Radke's observations which show a peak developing near 0.1 micron can be reasonably reproduced using a theoretical model which describes the coagulation of spheres. Fig. 1 shows the results of Radke et al. together with a theoretical calculation that did not assume any smoke spreading or dilution of the smoke plume. This assumption should provide the maximum rates of coagulation. As shown there, by 30 minutes a mode has developed at approximately the correct size, but the small particles measured by Radke et al. are not consistent with the losses calculated here by coagulation. This inconsistency remains even when the modeled smoke is diluted at rates of $4.6 \times 10^{-5}$ $s^{-1}$ (see Fig. 2).

Fig. 3 shows a similar calculation based on data taken by Hallett et al. (1986). In this case, a loss rate for the smoke of $1.0 \times 10^{-5}$ $s^{-1}$ was assumed. This loss rate was chosen to fit the observed loss of particles to the aging chamber walls once coagulation had essentially been quenched. For this experiment, the model calculation develops a mode near 0.06 micron while the data show that the mode develops near 0.2 micron. Also, as was observed for the Meteotron data, the depletion of the small particles by coagulation is not observed.

One possible explanation for the discrepancy between the model simulation of aging and the size distribution measured by Hallett et al. (1986) is that chained agglomerate and highly absorbing particles are not sized correctly by the instrument used to measure them in this experiment (R. Turco, private communication, 1986). The measurements of Hallett et al. were made with an ASASP instrument that is calibrated using non-absorbing spheres (W. Porch, private communication, 1986). Thus, it is not clear how it would size absorbing chains. The measurements of Radke et al. were taken using an electrical mobility analyzer (.01 to 0.1 micron radius) overlapped with a single particle light scattering instrument (0.3 micron to 10 micron). The particle light scattering instrument is also dependent on using non-absorbing spheres as a calibration. Also, the electrical mobility analyzer may routinely assign particle sizes that are smaller than their physical size based on their outlines in a scanning electron microscope picture (Hallett, private communication), so that this instrument too may not provide a good measure of particle size. We conclude that more analysis of the response of these instruments to known smoke particle sizes is needed before a good comparison to coagulation theory is possible.

3. Effects of aging on scavenging of smoke

If smoke particles are spherical and if they are chemically homogeneous, the largest particles will become nucleated at the lowest supersaturations. Because the largest particles in any smoke sample are formed by coagulation and because, as pointed out above, the confirmation of coagulation theory for smoke particles has not been straightforward, the interpretation of data pertaining to aging on the nucleation scavenging rates for smoke is difficult. However, we review here the recent data and suggest at least one consistent explanation.

Early experiments by Hallett et al. (1986) tested a number of laboratory-generated smoke samples as well as a few field samples of smoke for their ability to form cloud condensation nuclei (CCN). For acetylene smoke and for smoke from a field burn of diesel (40%) and gasoline (60%), they also aged the samples and reanalyzed the CCN characteristics. Both fuels produce a black, highly carbonaceous smoke. Typically, "fresh" smoke that has been diluted to concentrations ranging from $10^5$ to $10^6$ $cm^{-3}$, had a relatively small percentage of CCN. For example, the percentage of CCN active at 0.3% supersaturation for nearly fresh acetylene smoke ranged from 0.015% to 0.04% of the total number of particles. When the smoke sample was aged, the total number concentration of smoke particles decreased (due in part to coagulation and in part to wall loss in the aging chamber), but the percentage of particles that act as CCN increased. For the case of acetylene smoke active at 0.3% supersaturation, the percentage of particles which act as CCN increased to 2% after 55 minutes. In another experiment, acetylene smoke was aged for 16 hours. In this case after aging nearly 100 percent of the particles were active as CCN at 1% supersaturation.[2] Furthermore, the particles activated in this experiment had an apparent radius as measured by an electrical aerosol analyzer (EAA) of only 0.03 micron.

During smoke aging, the fraction of large particles will increase as the smaller particles coagulate and decrease in concentration. Thus, the aging experiments of Hallet et al. which show increasing fractions of CCN particles appear consistent with the theoretical expectation (derived for spherical smoke particles) that the largest particles within any distribution will activate at the lowest supersaturations, assuming, of course, that the particles have a homogeneous chemical composition. However, the finding that particles with a radius of only 0.03 micron can act as CCN is surprizing, in view of their high carbon content. Previous analysis had supposed that carbonaceous smoke particles would be hydrophobic (NRC, 1985). We note, however, that the size assigned by the electrical aerosol analyzer may be smaller than the actual physical size. More recent experiments by Hallett, in which acetylene smoke particles are first sorted by size and size is also measured by scanning electron microscope pictures of a filter sample, confirm the conclusion that very small smoke particles (with physical dimensions corresponding to radii ranging from 0.06 to 0.1 micron) can act as CCN at supersaturations of about 0.2 percent (Hallett, private communication, 1986).

[2] This conclusion assumes that the electrical aerosol analyzer (which cannot detect particles less than about 0.01 micron in radius) measured the entire distribution. However, it is unlikely that particles less than 0.01 micron would have survived coagulation over this time.

To illustrate the effects of aging on CCN and total particle concentration, Fig. 5 shows a calculation that simulates the one hour aging experiment for acetylene smoke of Hallett et al. (private communication, 1985). In this simulation, the initial distribution is assumed to be log-normal with a mean geometric standard deviation of 1.5 and a mode radius of 0.02 micron. Plotted are the total number of particles as well as the number of particles with radius greater than 0.1 micron. Also shown are the results from the aging experiment of Hallett. For this experiment, the time evolution of smoke particles, as well as the time evolution of CCN particles is reasonably consistent with a spherical model in which 0.1 micron particles will just activate at 0.3% supersaturation. Referring to Fig. 3, this would be consistent with the behavior associated with a particle whose mass fraction of soluble material was about 0.05. Of course, this nucleation behavior may not be due to the presence of any soluble material since nucleation may also be caused by surface processes and capillary action. However, it would be helpful to measure the chemical content of acetylene smoke particles, since the high flame temperature under which they form may have dissociated some molecular nitrogen which could have formed nitric acid and deposited on the particles.

Recall that the longer aging experiment with acetylene smoke (for 16 hours) resulted in a final CCN to CN ratio of close to 100 percent at 1% supersaturation. This behavior must be contrasted with that of an aging experiment for smoke from a field burn of diesel and gasoline fuel (Hallett et al., 1986). In that experiment, after an aging time of 54 hours, only 10 percent of the particles were active as CCN at 1% supersaturation. As discussed above, in this experiment a size distribution was also measured, using an ASASP optical probe. If this distribution is integrated from the largest size down and matched to the observed CCN concentration and if it is assumed that the largest particles were those which acted as CCN, then those particles with radii larger than 0.06 to 0.07 microns are activated at 1% supersaturation. Unfortunately, the particles from this fire are undoubtably chained agglomerates and, as pointed out above, the ASASP instrument may not size them properly. Furthermore, we have no separate sizing information (as is available for the acetylene smoke and the electrostatic classifier instrument) with which to calibrate the ASASP sizes. We note, however, that if the initial sample is dilute enough, the coagulation process may simply not proceed fast enough to convert a large percentage of the sample into CCN.

To illustrate the effect of initial particle concentration on the ratio of CCN to total particle concentration with aging, Fig. 6 shows three model calculations similar to that shown in Fig. 5, but assuming initial concentrations of $3.4 \times 10^6, 3.4 \times 10^5$, and $4.9 \times 10^4$ $cm^{-3}$. The initial size distribution was log-normal with a mode radius of 0.02 micron and a mean geometric standard deviation of 1.5. The observed chamber wall loss of $1.0 \times 10^{-5}$ $s^{-1}$ was assumed. In the figure, the separate cases are labled as having initial mass concentrations of $2.4 \times 10^{-10}$, $2.4 \times 10^{-11}$, and $3.4 \times 10^{-12}$ g $cm^{-3}$, respectively. The total number of particles as well as the number of particles with radii greater than 0.08 micron are plotted. In the first case, nearly all the particles become larger than 0.08 micron within the first 10 hours. In the second case, about 10 percent of the particles grow to sizes greater than 0.08 micron by the end of the experiment. In the third case, when the initial concentrations are highly dilute, there is only a slight change in the ratio of particles greater than 0.08 micron to the total number of particles. Thus the final ratio

of CCN to total smoke particles can be highly dependent on the initial amount of dilution of the smoke sample.

The diesel/gasoline aging experiment was also modeled using the observed initial size distribution (see Fig. 3). After 20 hours, the number of particles with radii greater than 0.08 micron agrees well with the observed CCN behavior, but, apparantly, the large number of CCN active before 20 hours are smaller than 0.08 micron. This suggests that the initial smoke distribution may not be chemically homogeneous (although other explanations may be possible). Apparantly, a significant fraction of very small particles act as CCN initially. During aging they would attach themselves to larger particles. It is conceivable that even after 54 hours of aging some of the smaller particles may still act as CCN while larger particles do not. Further experiments are needed in order to fully delineate what these data mean.

To summarize, it appears that for smoke from acetylene flames, very small particles can act as CCN at supersaturations of close to and even below 1%. For smoke from gasoline and diesel fires, most particles above 0.1 micron will probably activate at 1% supersaturation but there is some indication that smaller particles may also activate. On short time scales (e.g. below cloud base above a large, intense fire) rapid coagulation to sizes that would act as CCN, would require high initial smoke densities and restricted mixing in order to produce a distribution in which nearly all of the particles were large enough to act as CCN before reaching cloud base. Very high number concentrations are consistent with the coagulation-smoke plume model of Penner and Porch (1986) if initial size distributions peak near 0.01 micron, but their dispersion calculations would not allow coagulation to be that rapid. More realistically if concentrations peak near 0.05 micron, we estimate that about 50 percent of the particles would coagulate to a scavengable size below cloud base. In such a case, saturation ratios above a large, intense fire may be high enough that these particles would indeed activate (Penner and Edwards, 1986). We note that preliminary analysis of the residue of smoke particles from a rain-stained Hiroshima wall is also consistent with particles as small as 0.1 micron radius being scavenged in the firestorm that took place after the atomic bombing in August 1945.[3] (David Fields, private communication, 1986.) On longer time scales, if the particles grow to sizes near 0.4 micron in radius, nearly all of them may be large enough to act as CCN (Penner and Porch, 1986).

## 4. Conclusions

We have reviewed data and model simulations on the aging of smoke by coagulation. Smoke aging after a nuclear war may produce particles as large as 0.4 micron radius after a few days time. If the particles remain as chained agglomerates, the volume-equivalent radius of gyration for such a particle would be 2.4 micron. Data pertaining to the absorption and scattering of such large agglomerates is not available, although at least one experiment suggests that large particles may not aborb radiation as efficiently as smaller "fresh" smoke particles. Further experiments are needed to prove this hypothesis.

[3] Unfortunately, these data do not allow us to say what fraction of the total distribution was scavenged.

We have also reviewed data pertaining to smoke aging and its effect on CCN concentrations. Apparently, very small particles (with radii near 0.08 micron) can activate at supersaturations of close to 1 percent. Based on the results of Penner and Porch (1986), approximately half of the smoke particles would be larger than this before reaching cloud base above a large, intense fire. After coagulation for several days, nearly all of the particles could be large enough to act as CCN.

## 5. Acknowledgments.

This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract W-7405-Eng-48 and funded in part by the Defense Nuclear Agency.

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## Figure Captions

Fig. 1. Size distribution of smoke particles resulting from a calculation of smoke coagulation in the Meteotron plume. The calculation used the observed distribution at 18 minutes (Radke et al., 1980) and did not account for any spreading or dilution of the smoke plume. The symbols $\nabla$, $\Xi$, and + show the observed distribution at 18 minutes, at 30 minutes and at 40 minutes, respectively.

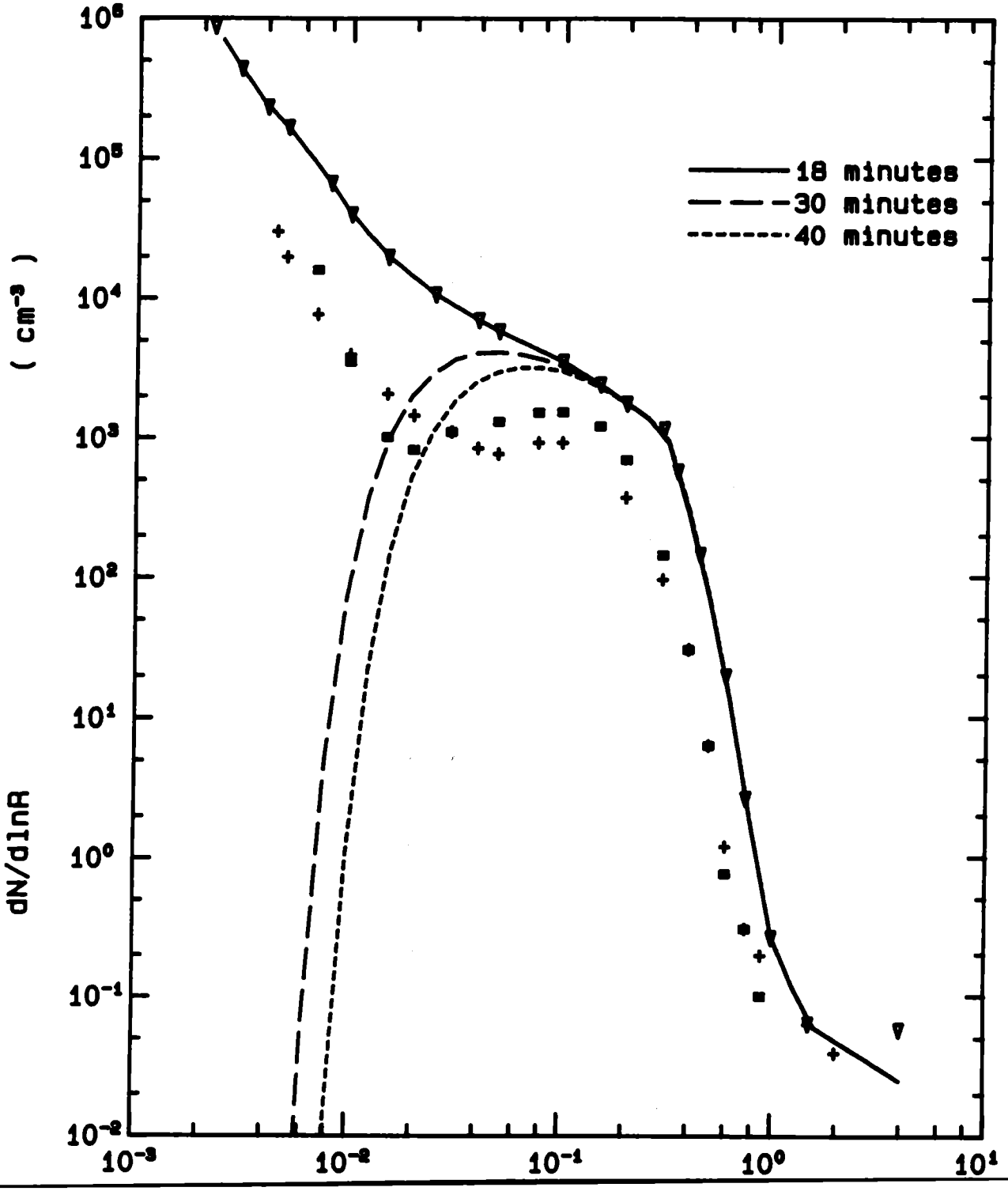
Fig. 2. Size distribution of smoke particles resulting from a calculation of smoke coagulation in the Meteotron plume. The calculation assumed a smoke dispersion rate of $4.6 \times 10^{-5}$ $s^{-1}$. Symbols are as in Fig. 1.

Fig. 3. Size distribution of smoke particles resulting from a calculation of smoke coagulation for the diesel/gasoline fire on Oct. 23, 1985 reported by Hallett et al. (1986). The initial concentration was fit to the observed distribution after two hours (symbol $\nabla$), and a loss rate for smoke adhesion to the chamber walls of $1.0 \times 10^{-5}$ $s^{-1}$ was assumed. The symbols $\nabla$, $\Xi$, and + refer to measurements taken after 2 hours, after 30 hours, and after 54 hours, respectively.

Fig. 4. Critical supersaturation as a function of particle radius for particles with a mass fraction of 0.0, 0.01, 0.1 and 1.0 of ammonium sulfate.

Fig. 5. Total particle concentration and concentration of particles with radii greater than 0.1 micron resulting from the coagulation of an initial log-normal distribution with a mode radius of 0.02 micron and geometric standard deviation of 1.5. The symbols refer to measurements of Hallett et al. (private communication, 1985) for total particle concentration ($\odot$) and CCN active at 0.3% supersaturation ($\nabla$).

Fig. 6. Total particle concentration (upper curves) and concentration of particles with radii greater than 0.08 micron (lower curves) resulting from the coagulation of an initial log-normal distribution with a mode radius of 0.02 micron and geometric standard deviation of 1.5. The symbols refer to measurements of Hallett et al. (1986) for smoke from a diesel/gasoline fire. The symbol $\odot$ refers to the measured total particle concentration while $\nabla$ refers to the CCN active at 1%. Also shown is the total particle concentration resulting from a calculation which was integrated using the observed initial distribution (see Fig. 3) (▣) and the concentration of particles greater than 0.08 micron (+).


18 minutes
30 minutes
40 minutes
dN/dlnR
( cm⁻³ )
10⁶
10⁵
10⁴
10³
10²
10¹
10⁰
10⁻¹
10⁻²
10⁻³
10⁻²
10⁻¹
10⁰
10¹

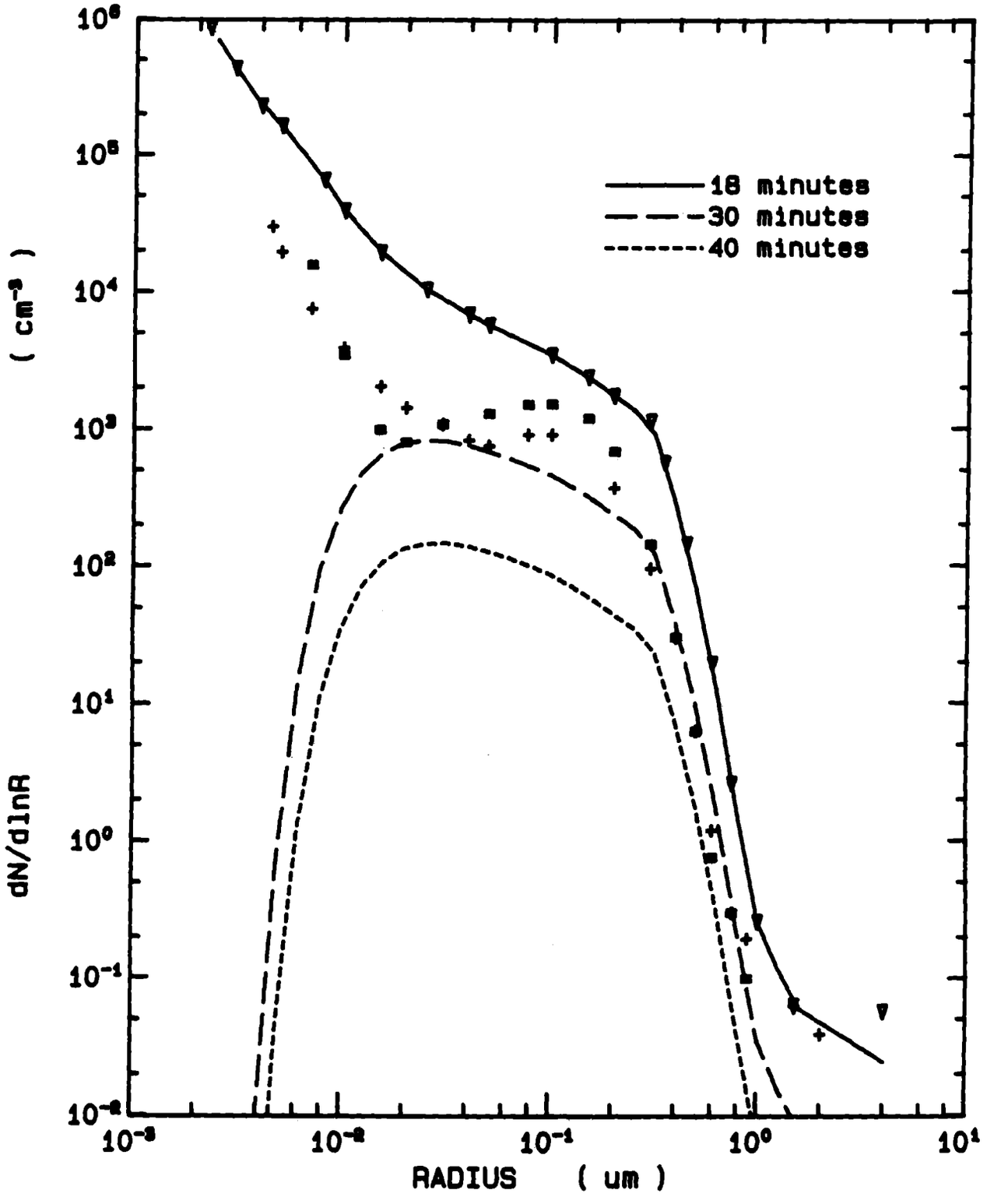

18 minutes
30 minutes
40 minutes
dN/dlnR ( cm⁻³ )
RADIUS ( um )
10⁶
10⁵
10⁴
10³
10²
10¹
10⁰
10⁻¹
10⁻²
10⁻³
10⁻²
10⁻¹
10⁰
10¹

Figure 2

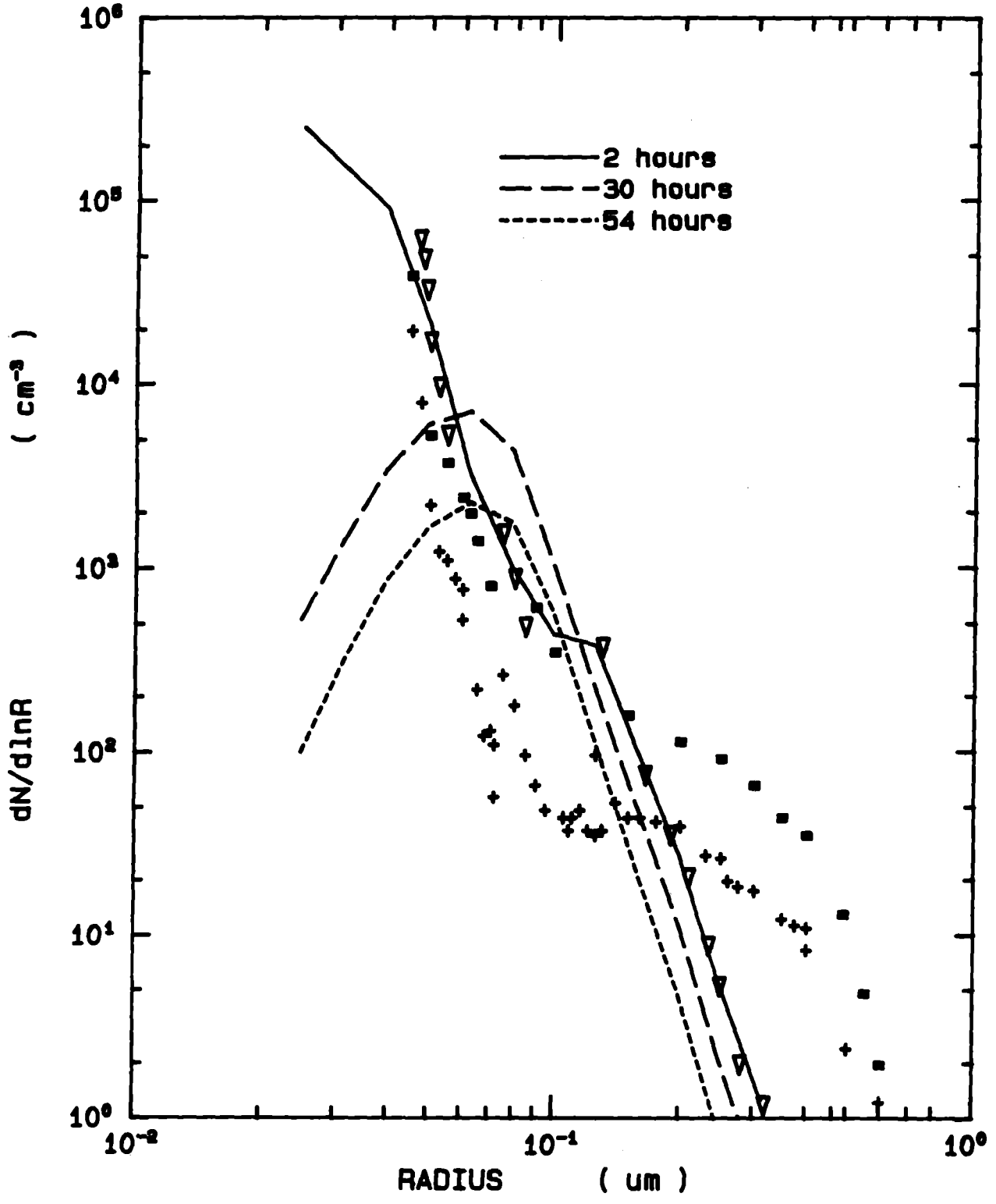

2 hours
30 hours
54 hours
dN/dlnR ( cm⁻³ )
RADIUS ( um )
10⁶
10⁵
10⁴
10³
10²
10¹
10⁰
10⁻²
10⁻¹
10⁰

Figure 3

# Critical supersaturation:

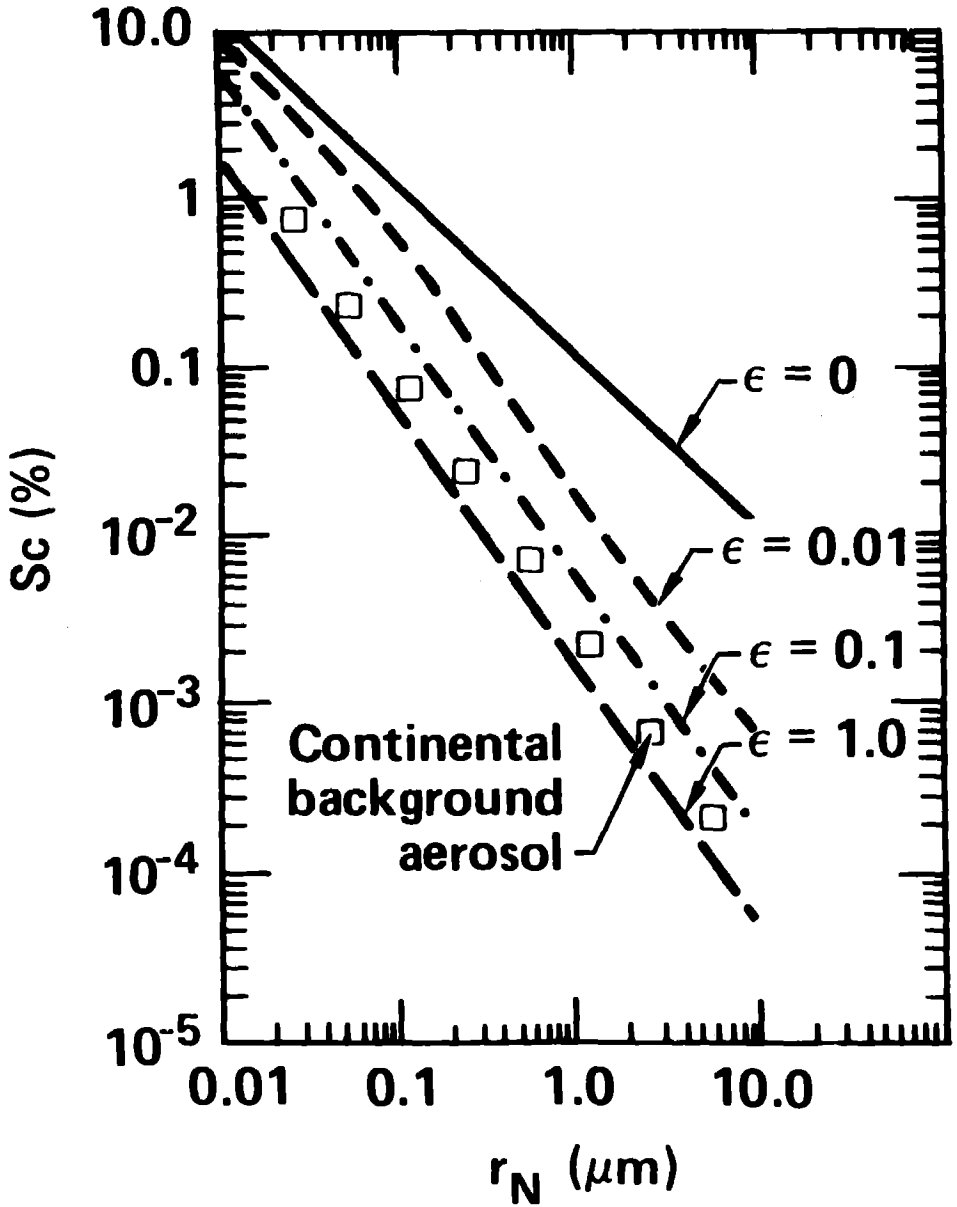



Figure 4

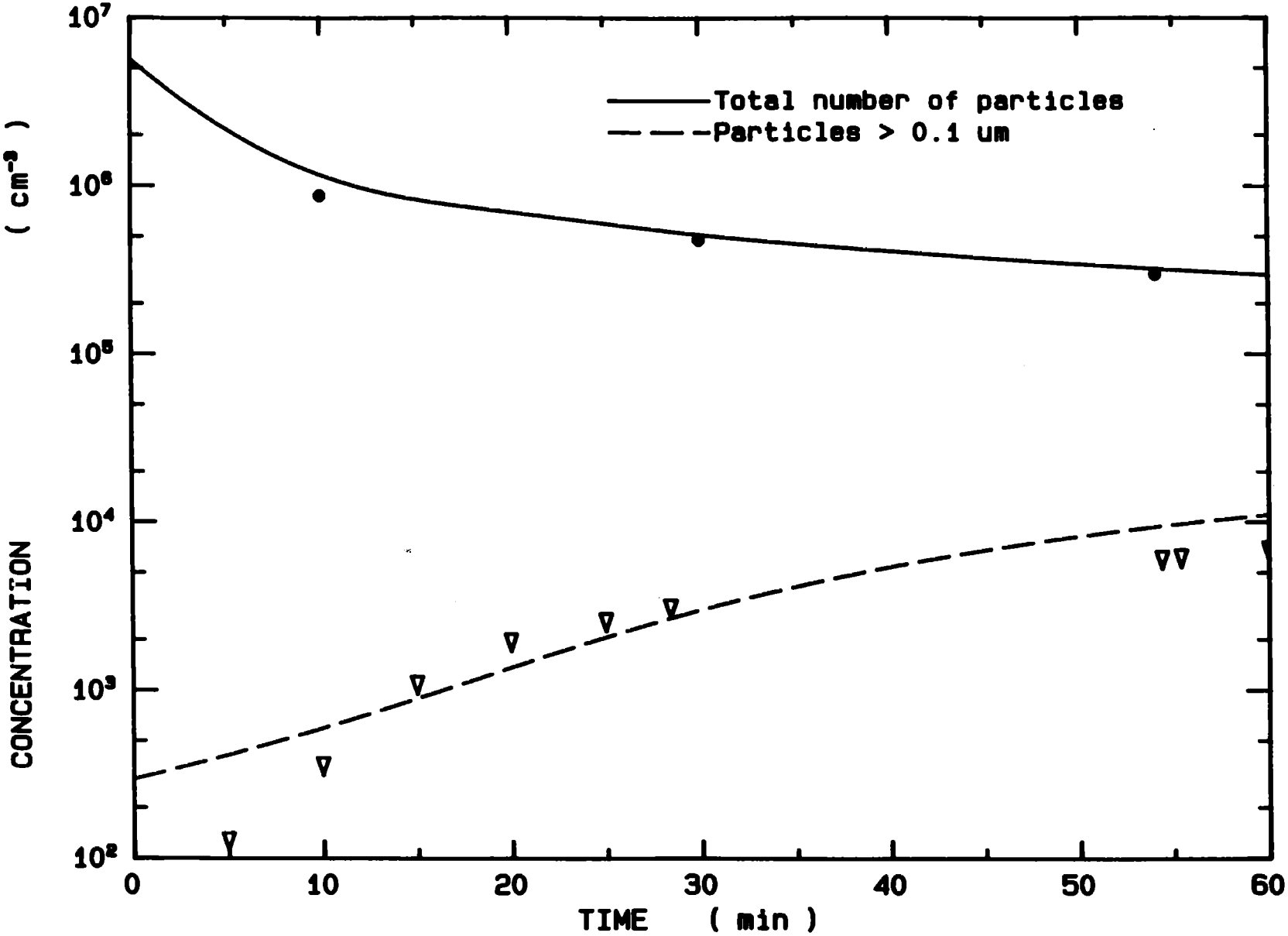

Total number of particles
Particles > 0.1 um
CONCENTRATION ( cm⁻³ )
TIME ( min )
10⁷
10⁶
10⁵
10⁴
10³
10²
0
10
20
30
40
50
60

Figure 5

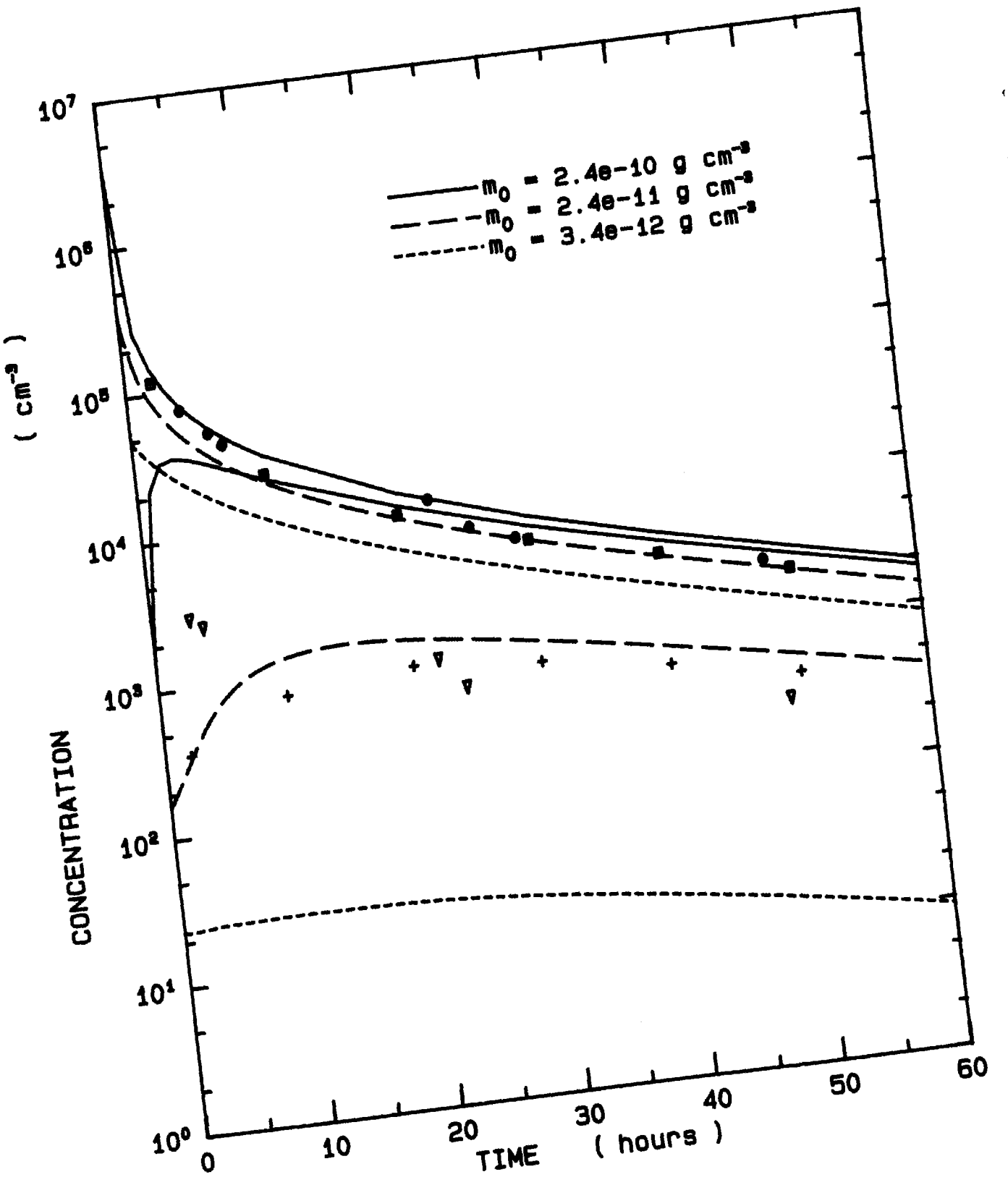

$m_0$ = 2.4e-10 g cm$^{-3}$
$m_0$ = 2.4e-11 g cm$^{-3}$
$m_0$ = 3.4e-12 g cm$^{-3}$
CONCENTRATION ( cm$^{-3}$ )
TIME ( hours )
$10^7$
$10^6$
$10^5$
$10^4$
$10^3$
$10^2$
$10^1$
$10^0$
0
10
20
30
40
50
60

Figure 6